UNIVERSITI PUTRA MALAYSIA

PREPARATION AND EVALUATION OF REDUCED GRAPHITE OXIDE FOR OIL SPILL REMOVAL

INTESAR RAZAQ HUSSAIN

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By

INTESAR RAZAQ HUSSAIN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirements for the Degree of Master of Science

September 2016

DEDICATION

With gratitude for their love, support, and guidance:
I dedicate this thesis to my beloved family and friends with love.

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the Degree of Master of Science

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September 2016

Chairman : Dayang Radiah Binti Awang Biak,PhD
Faculty : Engineering

The production of reduced graphite oxide, graphite oxide and their associates has sparked the interests of the nanomaterials scientists and engineers for the past decade. With their superior properties, these materials can be functionalized or modified to fit specific applications from nanocomposites to smart and functionalized materials. The typical and most efficient reduction agent used is Hydrazine Hydrate (N_2H_4), a highly toxic chemical. The objectives of this work were to synthesize and characterize the hierarchical and porous structure of reduced graphite oxide using reduction integrated chemical treatment methods. In addition, this study aimed to evaluate the potential of reduced graphite oxide as an absorbent for oil spill removal.

Graphite Oxide was synthesized using the modified Staudenmaier's method and chemically reduced using 30% heated Ammonia solution (NH_3) at 90˚C. Different chemical treatments were done namely leavening method (10 hours) and spaced method (5, 10 and 17 hours using either soxhlet or rotary evaporator). A two-step reduction (calcination of reduced graphite oxide (4 hours) under a nitrogen atmosphere at 500^{o}C) was also done. For the oil removal test, crude oil was recovered from a simulated sample and the absorbent was regenerated by washing it in hexane. The GO and rGO products were characterized using different characteristic tools.

In general, it was found that different methods produced reduced graphite oxide with different properties even though the same reducing agent was used throughout the experiment. The results showed that rGO paper lost most of O-H functional groups, less defects and more thermally stable compared to GO. The percentage of impurities was significantly reduced from 15.37 for graphite oxide to 0.23 for calcined reduced graphite oxide. All the reduced graphite oxide papers produced in this study had a density that was lighter than water. They are also hydrophobic and super oleophilic with a contact angle ~ 120^o. The produced materials were able to absorb oil up to 41.3 g g^{-1}, while completely repelling water. The reusability test for

the reduced graphite oxide paper was conducted for five cycles. It was found that the reduction in the absorption efficiency was from 3.5% to 4% after each cycle. This indicates that the paper could be used more than five times in the oil removal process. In addition, the efficiency of the COD removal was 95%.

This work had demonstrated effective methods to prepare the reduced graphite oxide paper. It offers a new alternative methods using either direct contact with ammonia vapor only or with both ammonia liquid and vapor, at a much lower temperature (~ 90˚C) and can be produced at ambient pressure. The material also showed a strong potential as an absorbent for oil spill removal.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Master Sains

PENYEDIAAN DAN PENILAIAN GRAFIT KURANG OKSIDA UNTUK PENYINGKIRAN TUMPAHAN MINYAK

Oleh

INTESAR RAZAQ HUSSAIN

September 2016

Pengerusi :Dayang Radiah Binti Awang Biak,PhD
Fakulti : Kejuruteraan

Penghasilan grafit kurang oksida, grafit oksida dan sekutunya telah mencetuskan minat saintis dan jurutera bahan nano semenjak sedekad yang lalu. Dengan ciri-ciri yang lebih baik, bahan-bahan ini boleh terus difungsikan atau diubah suai untuk memenuhi aplikasi tertentu seperti dari bahan komposit nano hingga bahan pintar dan memfungsi. Agen yang lazim digunakan dalam proses pengurangan adalah Hidrazina Hidrat (N_2H_4), sejenis bahan kimia yang sangat tinggi toksiknya. Objektif kajian ini adalah untuk mensintesis dan menciri grafit kurang oksida yang berstruktur hierarki dan poros menggunakan kaedah integrasi pengurangan - rawatan kimia. Selain itu, kajian ini juga menilai potensi grafit kurang oksida sebagai bahan penyerap dalam proses penyingkiran tumpahan minyak.

Grafit Oksida telah disintesiskan menggunakan kaedah Staudenmaier yang telah diubahsuai dan melalui proses pengurangan kimia menggunakan larutan Ammonia (NH_3) (30%) yang dipanaskan pada suhu 90˚C. Rawatan kimia yang berbeza telah dijalankan iaitu kaedah penaikan (10 jam), kaedah terjarak menggunakan air (5, 10 dan 17 jam di dalam soxhlet atau penyejat berputar). Kaedah pengurangan dua lamgkah (pengkalsinan grafit kurang oksida (4 jam) di dalam suasana nitrogen pada suhu 500˚C juga dilaksanakan). Untuk ujian penyingkiran minyak, minyak mentah telah dipulihkan daripada sampel simulasi dan penyerap dijana semula menggunakan heksana. Produk GO dan rGO telah dicirikan dengan menggunakan kaedah pencirian yang berbeza.

Secara umumnya, kaedah yang berbeza telah menghasilkan grafit kurang oksida yang mempunyai ciri-ciri yang berbeza walaupun agen penurunan yang sama telah digunakan dalam kesemua eksperimen. Secara umumnya, ia kertas rGO yang dihasilkan kehilangan kumpulan fungsi O-H, kurang kerosakannya, dan lebih stabil sifat termanya. Menariknya, peratusan bendasing telah berkurangan daripada 15.37 dalam grafit oksida kepada 0.23 dalam grafit kurang oksida dikalsin. Semua kertas grafit kurang oksida dihasilkan dalam kajian ini kurang tumpat dibandingkan dengan air. Ia juga bersifat hidrofobik dan superoleofilik dengan sudut kontak

sekitar 120^{o}. Produk yang dihasilkan mampu menyerap minyak sehingga 41.3 g g^{-1}, dan menangkis air sepenuhnya. Ujian guna semula untuk kertas grafit kurang oksida telah dijalankan untuk lima kitaran. Pengurangan kecekapan penyerapan hanya 3.5 hingga 4% sahaja selepas setiap kitaran. Ini menunjukkan kertas ini boleh digunakan dengan cekap lebih daripada lima kali bagi proses penyingkiran tumpahan minyak. Di samping itu, kecekapan penyingkiran COD adalah 95%.

Kajian ini telah menunjukkan satu kaedah efektif untuk menyediakan kertas grafit kurang oksida. Ia menawarkan kaedah alternatif baharu menggunakan sentuhan terus wap ammonia sahaja atau dengan wap dan larutan ammonia pada suhu yang lebih rendah dan boleh dihasilkan pada tekanan ambien. Produk ini juga menunjukkan potensi yang baik sebagai penyerap untuk penyingkiran tumpahan minyak.

ACKNOWLEDGEMENTS

I am deeply grateful to Allah S.W.T for giving me the strength and patience to enable me to complete this thesis.

The work presented in this study was conducted at Chemistry Department, Faculty of Engineering, University of Putra Malaysia (UPM). Firstly, I would like to especially express my appreciation to my research advisor, Dr.Dayang Radiah Binti Awang Biak, who not only served as my supervisor but also encouraged and assisted in the completion of this thesis. Without her help and encouragement, it would be impossible for me to begin my study. I am also grateful to the other committee member, Dr. Faizah bt. Mohd Yasin and Dr. Suryani bt. Kamarudin.

In addition, I would like to thank my friends and classmates in UPM who have expressed interest in my work and given me a lot of motivation.

Finally, I would like to thank my family members for their support during these years of my studies. I would especially thank my father who has inspired me through the years and my mother who has helped me immensely during my studies.

I certify that a Thesis Examination Committee has met on 30 September 2016 to conduct the final examination of Intesar Razaq Hussain on his thesis entitled "Preparation and Evaluation of Reduced Graphite Oxide for Oil Spill Removal" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

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I hereby confirm that:

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LIST OF ABBREVIATIONS

ASTM	American Society for Testing and Materials
BET	Brunauer, Emmett and Teller
EDX	Energy dispersive X-ray
FTIR	Fourier Transform Infrared Spectroscopy
eGO	Graphene oxide
GO	Graphite oxide
ITOPF	International Tanker Owners Pollution Federation
O/C	Oxygen to Carbon ratio
rGO	Reduced Graphite Oxide
reGO	Reduced Graphene Oxide
SEM	Scanning electron microscopy
SWCNT	Single-wall carbon nanotube
TEM	Transmission electron microscopy
TGA	Thermogravimetric analysis
UV-vis	Ultraviolet–visible spectroscopy

CHAPTER 1

INTRODUCTION

1.1 Background of Research

Nanotechnology has had a significant impact on the development of novel environmental engineering processes and technologies. This impact has been reflected in the benefits of using nanotechnology to deal with environmental pollution, predicting and preventing future environmental problems (Karn et al., 2009). Water pollution has become one of the most serious environmental challenges that human beings are facing. The application of nanotechnology for water treatment and quality control is expected to become a major industry over the next 10 to 15 years (Shan, 2010). Pollution by the petroleum industry and oil transportation causes severe environmental and ecological problems every year (Wang and Geng, 2015). The collection and recovery of oil spills using absorbents are promising methods which have attracted serious attention due to their high clean-up efficiency (Yang et al., 2014). The ideal materials for efficient adsorption of oil should have a high specific surface area, with inherently hydrophobic nature and excellent chemical stability (Yang et al. 2014). Among the absorbents available, carbon-based materials are suitable candidates for oil absorption (Chen et al., 2015).

Graphite oxide (GO) was first prepared almost 150 years ago by Brodie who treated graphite repeatedly with potassium chlorate and nitric acid (Zheng & Kim, 2015). Hummers and Offeman in 1958 demonstrated a less hazardous and more efficient method for graphite oxidation by involving a mixture of sodium nitrate, potassium permanganate and sulfuric acid. Their modified protocol is presently the most commonly used method for the oxidation of graphite (Eigler, 2011). The term ‘‘sheets’’ usually indicates monolayer to several layers whereas the term ‘‘platelets’’ is often used to describe thick multilayer (Zheng et al., 2014).

GO can partially reduce to produce reduced graphite oxide (rGO) sheets by removing the oxygen-containing groups (Petridis et al., 2016). Until now, the most promising way to produce rGO sheets is the chemical method (Park et al., 2008). Although, the material derived from this method usually consists of a significant amount of oxygen functional groups and defects, it is simple, not expensive and suitable for a broad range of mass production (Pei & Cheng, 2012). In most instances, during the assembly method, the massive accessible surface area of two-dimensional (2D) rGO sheets is lost. This situation is due to the individual rGO sheets that can irreversibly aggregate and restack due to π-π stacking and van der Waals forces between the planar basal planes of rGO sheets (Niu et al., 2012). In fact, this situation affects the potential applications of these materials in composite, electrochemical electrode and *etc*. (Loryuenyong et al., 2013).

In recent years, there has been a growing interest in the integration of 2D nanoscale building blocks, like rGO sheets, into three dimensional (3D) macroscopic structures (Schwinté & Keller, 2015; Lee et al., 2015; Zhang et al., 2016). These structures are drawing a lot of attention; therefore, there is a need to properly explore the advanced properties of individual 2D sheets for various applications (Geim & Novoselov, 2007). For example, free-standing rGO macroscopic structures have demonstrated unique catalytic, mechanical, and electrochemical properties with the potential applications as chemical filters and electrodes for energy storage devices (Chen et al., 2013).

Zheng et al., (2014) observed that exfoliated graphite oxide (eGO) sheets become hydrophobic and aggregated instantly in the solution when underwent the reduction process with the removal of oxygenated groups. This property is in contrast with that exhibited by the 3D structures. The 3D reduced exfoliated graphite oxide (reGO) structure displayed sorbtion capabilities which reveal the potential application in the cleaning of pollutants where the recovery, reusability, and regenerability of the sorbent are important (Shan, 2010). In recent years, nanotechnology has emerged as a possible source of novel solutions for several of the world's pollution problems. However, the application of nanotechnology for oil spill cleanup is still in its infancy stage (Mahajan, 2011), but it offers great promise in the long run. Within the last few years, there has been notably growing interest worldwide in exploring ways to find suitable solutions to clean up oil spills through the utilization of nanomaterials (Mahajan, 2011). The major drawbacks are mainly concerning the challenges in re-collecting the nanomaterials when they are found in powder forms.

The ideal material for efficient sorption of oil should have a high specific surface area. The material should show a hydrophobic nature with high chemical stability. In this context, carbon-based materials are among the best candidates for oil sorption. Currently, several kinds of carbon-based material, like activated charcoal, and expanded carbon nanotubes (CNTs) had been developed and tested to remove crude oil product from water (Tan et al., 2008; Nardecchia et al., 2013). The 3D frameworks, e.g. aerosols, foams and sponges are the new types of solid macro porous nanomaterials with interconnected micro porous structures, low density and large surface area that are developed and have the potentials as effective sorbents. The 3-D materials have both hydrophobic and oleophilic properties; therefore, they attract a lot of attention as efficient oil sorbents (Yang et al., 2014).

1.2 Application of Reduced Graphite Oxide in Water Pollution Control

Oil pollution is probably one of the most important subjects in any discussion of environmental pollution. The water we drink, the air we breathe, the soil in which our crops are grown, and the environment in which animals and plant grow are continually contaminated by a variety of synthetic chemicals (Saravi & Shokrzadeh, 2011). The environmental contamination by petroleum and its derivatives is a problem of increasing magnitude with obvious ecological and economic implications.

Oil spills result in the loss of a valuable source of energy. Therefore, the efficient way to recover and utilize the spills is of great research interest. Oil spills from tankers that transport 60 % of the world's oil are a serious threat to the environment when several traffic routes cross the boundaries of various marine ecosystems and marine hot spots (Yang et al., 2014).

At the point when oil comes in contact with water, it forms on oil-in-water emulsion or or floating film that should be removed before the oil is dispersed to a much larger area (Keshawy et al., 2013). Even low concentrations of oils can be toxic to microorganisms that are responsible for bio-degradation in a typical waste water process. Another threat to the environment comes from polycyclic aromatic hydrocarbons (PAHs) that are known to have an effect on a range of biological processes and may be potent cell mutagens and carcinogens (Das, 2011).

Previous studies have reported on different methods for oil spill cleanup such as oil-sorption materials, in situ burning, mechanical recovery, physical diffusion, enhanced bioremediation, oil skimmers, and superhydrophobic materials (Doerffer, 2013). Among these methods, oil–water separating materials with both superhydrophobic and superoleophilic properties have attracted a lot of attention in both fundamental research and potential application in the field of oil–water separation (Chen et al., 2015). Due to the complete removal of the oil from the oil spill site and high possibility of re-collection, these materials can sometimes be recycled (Yang et al., 2014). Recently, various superhydrophobic and superoleophilic materials such as carbon nanotubes, graphene zeolites and organo-clay have been investigated for their ability to separate the oil-water mixture (Xi et al., 2014). For example, modified organophilic clay, lime, silicas, exfoliated graphite, plastic polymers, cellulose-based materials and elastomers (Carmody et al., 2007; Meng & Park, 2012). All of these materials showed high porosity and the ability to absorb oil from water.

Oil-sorbent materials can be classified into three major classes; namely, inorganic mineral products, organic (plant-based) products and synthetic organic products (Behnood et al., 2013). Mineral products include materials such as zeolites, silicas, perlites, graphite, vermiculites, sorbent clay, and diatomite (Likon et al., 2011). Organic (plant-based) products (or natural sorbents) include straws, corn cobs, wood fibers, cotton fibers, cellulosic kapok fibers, kenafs, milkweed floss and peat moss (Ongarbayev & Belgibayeva, 2015). Such organic (plant-based) products showed poor buoyancy characteristics, relatively low oil sorption capacity and low hydrophobicity (Ying et al., 2014). Synthetic organic products include polymeric materials such as polypropylene and polyurethane foams, and graphene foams which are the most commonly used commercial sorbents in oil spill cleanup due to their oleophilic and hydrophobic characteristics (Das, 2011; Jiang et al., 2015; Niu et al., 2012 ; Zhang et al., 2016).

The sorbent chemical water mixture can be disposed mechanically from the water surface. Like all other cleaning up principles, sorbents can only work at the water surface; therefore, the bulk density of the sorbents must be less than the density of water. The sorbents then can sink into the chemicals to be wetted totally. Sorbents can work through the mechanism of absorption, adsorption, or both as sorption (Iqbal & Abdala, 2013).

1.3 Statement of The Problem

The chemical method has become a promising way to produce rGO. The material derived by this method contains a significant amount of oxygen functional groups and defects. This method is simple, inexpensive and suitable for large-scale or mass production. The process involves graphite oxidation and followed by the reduction of GO sheets.

Most reduction methods utilized strong reduction agents such as hydrazine (Chua & Pumera, 2016). Works on reduction process utilized ammonia solution involved in using eGO as a source and depend mainly on the direct liquid contact (contact with the solution) (Zhuo et al., 2015; Liu et al., 2015; Pham et al., 2013). Their application were in the electrochemical sector.

Furthermore, revious research had mainly focused on 2D constructs. The important technological challenge is the restacking of the 2D structure which strongly reduces the active surface area of rGO. The restacking is likely caused by the strong sheet to sheet van der Waals interactions (El-kady et al., 2012). Therefore, to take full advantage of the large surface area of rGO and chemical functionality, 2D rGO must be integrated into the macroscopic 3D structure to prevent such restacking (Byon et al., 2011; Worsley et al., 2012; Worsley et al., 2010).

The integration of individual rGO sheets into 3D macroscopic assembly and then into a functional system is a major method for macroscopic applications. Constructing 3D rGO based structure has become one of the most active research fields in the past few years (Nardecchia et al., 2013; Min et al., 2013). In recent studies, a large number of novel methods have been developed to prevent restacking such as Vertically Oriented Graphene Sheets (Miller, 2012), Deformed Graphene Sheets (Liu et al., 2010; Liu et al., 2012; Wen et al., 2012), Spaced Graphene (Si and Samulski, 2008; Wang et al., 2011; Fan et al., 2010), 3D Graphene Networks (Worsley et al., 2012; Sheng et al., 2012; Li et al., 2013; Li et al., 2012). Furthermore there are numerous reports in literature suggesting possible applications of rGO and their derivatives due to its attractive characteristics (Gao et al., 2011, Kysilka et al., 2011, Liu et al., 2011, Min et al., 2011, Pang et al., 2011, Sheng et al., 2011, Guixia et al., 2011).

Crude oil has become the foremost vital energy resource for human beings. With the increasing numbers of crude oil fields, transportation, storage of crude oil and its products, the risk of oil spills is also increasing, both in marine areas and fresh water. Between 1974 and 1994, there had been 175 major oil spills worldwide. It had cost as high as from $20 to $200 per liter to clean up the contaminated sites, depending on the location and type of oil spill (Abdullah et al., 2010). In addition, the oil spills from transport by tank ships, pipeline, rail, and truck or from production activities were also serious. Between 1998 to 2007, about 11,000 tons of pipeline spills about 1,700 tons of refinery spills, 1,300 tons of tanker truck spills, and 500 tons of tank ship spills were reported to occur annually in the United States alone (Xiao, 2012). Therefore, the need for cost-effective technologies to deal with oil-water separation has become urgent in light of the forguest oil spill events.

This study utilized GO as a potential absorbent for oil spills. However, due to the hydrophilic nature of GO, this material cannot be directly used as it has to selectively and efficiently remove oil from water. Hence, it is necessary to eliminate its hydrophilicity properties. In view of this, chemical reduction was used to increase surface hydrophobicity as the possibility of restacking of GO during the chemical reduction process could affect the formation of high surface area. This problem was investigated in this study by applying various processing methods.

1.4 Objectives

The main aim of this study was to investigate the suitability of the preparation method to produce rGO paper at low temperture and ampedint condition. The prepared paper could be used as an oil absorbent. To achieve this aim, the following objectives were outlined:

1. To evaluate the effects of reduction process of ammonia solution to synthesize reduced graphite oxide.
2. To characterize the properties of the prepared rGO paper.
3. To investigate the potential of the rGO paper as oil absorbent.

1.5 Scope of The Study

The scopes of this study were clearly defined to achieve the goal of this study and are listed as follows:

1. RGO paper was prepared via two different methods involving two major steps; namely, the synthesis and chemical reduction steps. The first step was the synthesis of GO using Staudenmaier Method. The chemical reduction process was performed by exposing GO in ammonia solution (NH_3 solution). In this study, it was presumed that the substitution of NH_3 solution can be an effective agent for the reduction of GO.

2. The characterization of rGO morphology was done using Energy Dispersive X-ray (EDX) to check the ratio of oxygen to carbon. The Scanning Electron Microscopy (SEM) and Transmission Electron Microscope (TEM) produced the miscroscopic images of the samples. The Fourier Transform Infra-Red (FT-IR) tests detected the changes in the functional groups upon modifications. The Ultra Violet- Visible Light Spectrophotometer (UV-Vis) was used as a fast technique to check the formation of GO based on the wavelength. The thermal degradation of the samples was checked using Thermogravimetric Analysis (TGA) unit. Other analyses included the surface area and porosity.

3. The amount of oil adsorbed was measured by calculating the difference in the weight of the sorbent before and after it was subjected to the spill. The ability of the sorbent to retain the oil was measured by calculating the amount of oil held by the sorbent at a set time.

1.6 Thesis Outline

This thesis is organized into five chapters. Chapter 1 introduces the subject and presents the problem statement. Chapter 2 encompasses the reviews on the subject of this study, the analysis and synthesis of previous works which are related to GO, rGO paper with different reduction agents, and the various methods to clean up oil spills. Chapter 3 presents the types of materials, experimental set up, respective analytical equipment, and experimental approaches for this study. Chapter 4 presents the results and discussions on the obtained results. The quality of GO and rGO papers produced is highlighted and the best conditions for producing rGO papers to be used in the oil spill removal process are also discused. Chapter 5 concludes the findings and proposes recommendations for future works.

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